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LETTER TO THE EDITOR

Cross sections of the Kr II transitions at 96.5 and 91.7 nm excited by electron impact

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Abstract. The emission cross sections of the Kr II resonance transitions at 96.5 and 91.7 nm have been measured as $3.4 \times 10^{-18} \text{ cm}^2 \pm 14\%$ and $2.8 \times 10^{-18} \text{ cm}^2 \pm 14\%$ respectively at an incident electron energy of 200 eV. Their ratio was found to have a value of $1.22 \pm 12\%$ independent of electron energy over the range 100–300 eV. Spectral resolution and resonance trapping effects are shown to play a significant role in these measurements and may explain some of the wide divergence in previous results.

The ion transitions $ns^2np^5\ ^2P_{1/2,3/2} - nsnp^6\ ^2S_{1/2}$ excited by electron impact on the noble gases are of interest from both theoretical and practical points of view. The initial excitation process involves promotion of an inner-shell electron into the continuum leaving the resultant ion in an excited state. Because the ground state of the ion is a doublet, a pair of spectral lines result, for example the Kr II lines at 91.7 and 96.5 nm or the Ar II lines at 92.0 and 93.2 nm. The ratio of the cross sections for the two transitions provides useful information about the theoretical description of the excited species (Luyken *et al* 1972, Zapesochnyi *et al* 1974). The ion lines are unpolarised, since they originate in a state which is spherically symmetric and their excitation is unaffected by resonance trapping because of the very low ion density in the collision region of a typical electron impact experiment.

For these reasons the lines are potentially ideal candidates for use as secondary standards in the critical vacuum-ultraviolet (VUV) spectral region (90–100 nm) but, unfortunately, their excitation cross sections have not yet been established with sufficient accuracy to enable this to be accomplished (Forand *et al* 1988, van der Burgt *et al* 1989, Heddle and Gallagher 1989). In Ar there is good agreement on the intensity ratio ($1.86 \pm 10\%$) of the two lines even though considerably larger disagreement exists between the various measurements of the cross section for a particular line. However, as has been pointed out by Van Zyl (1988) the situation with the Kr lines is particularly bad with very significant discrepancies even in the ratio of the excitation cross sections of the two lines. The first determination of this cross section ratio was made by Luyken *et al* (1969) who obtained a value of 1.54. Later on, van Raan (1973) measured their absolute excitation cross sections at 300 eV with a ratio of 2.5. Luyken *et al* (1972) measured the excitation cross section of both lines combined by normalising to van Raan's data at 300 eV. Very recently, van der Burgt (1988) estimated a ratio of 1.22 from survey spectra taken at rather high source gas pressure, using the technique and apparatus described by McPherson *et al* (1986). In view of this wide spread it was felt worthwhile to make a careful remeasurement of the cross sections of the lines and

of their intensity ratio, paying particular attention to possible sources of error which might have been present in some of the earlier work. The present letter reports the results of this study.

The apparatus used in this work has been described in previous publications from this laboratory (Donaldson *et al* 1972, Malcolm *et al* 1979, Forand *et al* 1986) and, thus, only a brief description is given here.

The VUV emissions following electron impact on Kr were observed at right angles to a crossed electron-beam-gas-beam arrangement using a half metre Seya-Namioka VUV spectrometer with a gold-coated grating blazed at 70 nm (reciprocal linear dispersion 1.7 nm mm^{-1}). The photon detector used was a Mullard 419 BL channel electron multiplier. The electron current was controlled between 20–150 μA . In this region the photon counts were linearly proportional to the current. The gas beam was operated at a source pressure normally in the range 10–50 mTorr as monitored by a MKS Instruments capacitance manometer. A check was made to ensure that the output line intensities varied linearly with source pressure. Kr gas with a stated purity of better than 99.99% was used without further purification.

The calibration of the detection system in the wavelength range 90–130 nm has been performed in various ways in this laboratory (Forand *et al* 1988, Wang *et al* 1989). Basically, the known cross sections of the H, H₂, N₂ and rare gas transitions are used to establish calibration points on a relative scale. The Lyman- α emission cross section for dissociative excitation of H₂ (Woolsey *et al* 1986) is used for normalisation. In the spectral region of interest in this work, the errors involved in the calibration are estimated to be less than 15% in the absolute sensitivity of the detection equipment with an error in the relative sensitivity over the range 91–97 nm being much less than this.

The first concern in the determination of the cross sections is the proper isolation of the spectral lines of interest. The spectral data (Kelly and Palumbo 1973) clearly show a possibility of contamination of both the 96.5 and 91.7 nm lines by neighbouring features if insufficient spectral resolution is used. A Kr I line at 96.3 nm might contaminate Kr II 96.5 nm, while an unclassified Kr line at 91.8 nm could be contributing to the Kr II 91.7 nm intensity. No evaluation of the magnitude of these effects was included in any of the earlier works.

Figure 1 shows emission spectra taken under a variety of conditions and clearly illustrates the magnitude of spectral contamination which could occur. Figure 1(a), taken with a spectral resolution of 0.51 nm (FWHM), shows no sign of contamination of the 96.5 nm feature but significant overlap occurring at 91.7 nm. The higher resolution spectrum, figure 1(b), clearly resolves the structure near 91.7 nm but also demonstrates the weak presence of the underlying 96.3 nm neutral Kr line which was not at all evident in figure 1(a). It is clear from figures 1(a) and (b) that insufficient spectral resolution could lead to a completely erroneous estimate of the measured cross sections and thus of their ratio.

A related factor which could affect the measurements is the source pressure in the target region. The measured intensities of the neutral Kr emissions will be strongly pressure dependent in a non-linear way because of the strong trapping of this resonance radiation which occurs. The Kr⁺ emissions, on the other hand, are unaffected by this effect because of the low ground-state ion densities. This effect is illustrated in figure 2, which shows the variation of output intensity with source pressure for the 91.7 nm Kr II and the 94.6 nm Kr I features. The strong self-trapping of the latter line is clearly evident. Figure 1(c) shows a spectrum taken at relatively high source pressure. It

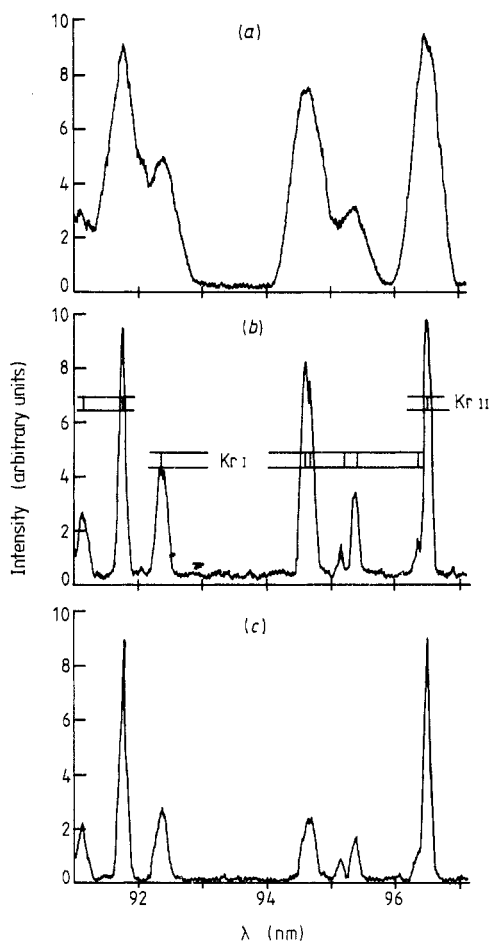


Figure 1. Emission spectrum of Kr following 200 eV electron impact: (a) with a spectral resolution of 0.51 nm at 70 mTorr gas pressure, (b) with a spectral resolution of 0.17 nm at 45 mTorr gas pressure, (c) with a spectral resolution of 0.17 nm at 170 mTorr gas pressure.

clearly demonstrates the reduced intensity of the Kr I lines relative to their Kr II neighbours. It is evident that a relative intensity measurement of the Kr II lines could be significantly pressure dependent if poor spectral resolution were employed. Clearly also, if poor spectral resolution is employed, it is likely that the most accurate value for the intensity ratio will be obtained at relatively high source pressure where the relative effect of the Kr I contributions would be minimised. This explains the agreement (table 1) between the result of van der Burgt and the present data. Since both Kr II lines originate from the same excited level ($4s4p^6\ ^2S_{1/2}$), the cascade population to this state should affect both lines equally. Therefore, their cross section ratio should be independent of electron impact energy, and represent the relative transition probabilities between the states involved. Measurements using different impact energies in the range 100–300 eV have confirmed this. Table 1 lists our result together with the previous data. The standard deviation of ten different measurements was less than 2%. The cross section ratios of Luyken *et al* (1969) and van Raan (1973) are 28% and 108% larger than the present result respectively. The 12% error quoted in our

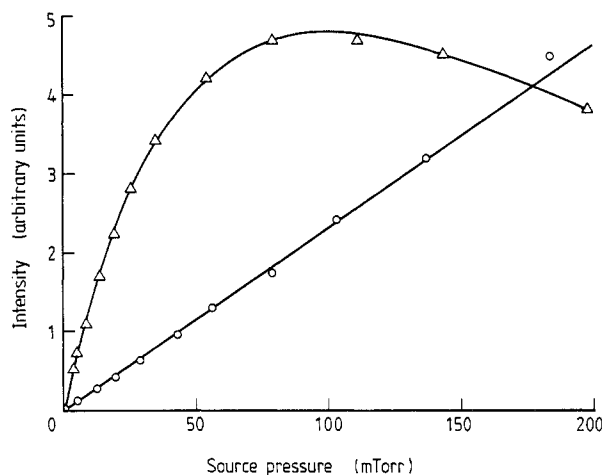


Figure 2. Emission line intensity against gas pressure curves at an incident electron energy of 200 eV: \circ , Kr II 91.7 nm line; \triangle , Kr I 94.6 nm doublet.

Table 1. 96.5/91.7 Kr II cross section ratio following electron impact.

Group	Ratio	Energy (eV)
Luyken <i>et al</i> (1969)	1.54	unknown
van Raan (1973)	2.5	300
van der Burgt (1988)	1.22	300
Present	$1.22 \pm 12\%$	100–300

result includes a possible 10% error in the relative wavelength sensitivity of our monochromator-detector combination over the range 91–97 nm.

Absolute values for the apparent excitation cross sections of the two lines were measured at a single electron energy (200 eV) by direct comparison of the emitted intensities with that of Lyman- α from H_2 excited under identical conditions, making use of the previously established variation of detector sensitivity with wavelength. The values obtained were 3.4×10^{-18} and $2.8 \times 10^{-18} \text{ cm}^2$ for the 96.5 and 91.7 nm lines respectively. The estimated error of $\pm 14\%$ is the root-square addition of the uncertainty in the relative wavelength sensitivity (10%), the statistical uncertainty in the count rates (2%), the error in the Lyman- α secondary standard (8%) and the current and pressure variations (each 2%). The sum of our two cross sections is 7% higher than the measurement of the two lines combined by Luyken *et al* (1972), a satisfactory but possibly fortuitous level of agreement considering the method of normalisation used by Luyken *et al*.

The cross section ratio of the Kr II resonance transitions at 96.5 and 91.7 nm excited by electron impact on Kr has been determined to be $1.22 \pm 12\%$ under single collision conditions over the energy range 100–300 eV. Absolute apparent excitation cross sections for the two lines were measured to be $3.4 \times 10^{-18} \text{ cm}^2$ and $2.8 \times 10^{-18} \text{ cm}^2$ respectively with an error of $\pm 14\%$ at an incident electron energy of 200 eV. Spectral resolution and resonance trapping are demonstrated to be major problems in the experiment. Together with the Ar II lines at 92 and 93.2 nm, these lines can now be

used with some confidence for relative spectral calibration in the critical 90–100 nm spectral region.

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